

Thermal and Elastic Properties of Thin Films

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Abstract—Optical, electronical and structural properties of thin films have been largely reported on literature. On the other hand, thermal properties have received little attention. Here we adopted two different techniques to determine thermal and elastic constants of thin films. The first was the bending beam technique, based on a deflection of a laser beam, used to determine the radius of curvature of the film/substrate composite, allowing extracting the stress of thin films. By making these measurements in substrates with different coefficient of thermal expansion we can also obtain a mixed elastic constant (biaxial modulus) involving a combination of the Young's modulus and the Poisson's ratio. Combining these results with a second technique, i.e., nanoindentation measurements, we demonstrate that one can obtain a total of 5 different thermal and elastic properties of thin films: stress, hardness, coefficient of thermal expansion, Young's modulus and Poisson's ratio. The procedure was used to obtain these parameters of hydrogenated diamond-like carbon (DLC) thin films.

Keywords—Coefficient of thermal expansion, Young's modulus, Poisson's ratio, stress, hardness.

I. INTRODUCTION

Mechanical and structural properties of amorphous carbon are largely reported in literature. However, there are few works reporting the thermal properties, such as the coefficient of thermal expansion (CTE) [1-4]. It is well known that amorphous carbon tends to have high stress [5,6] that hinders the measurements of properties that require relatively thick material, such as the measurement of thermomechanical properties. The high stress limits the production of stable thin films with thickness of few microns, which is necessary for the measurement to the CTE using the bending beam technique.

Here we report on a combination of the measurement of two properties, stress and nanoindentation (nanohardness), that allow the determination of 3 additional thermal and elastic properties of thin films, i.e., the coefficient of thermal expansion, Young's modulus and Poisson's ratio. This procedure was adopted to determine those parameters of amorphous hydrogenated diamond-like carbon (DLC), a-C:H.

II. EXPERIMENTAL

The DLC films were prepared by plasma enhanced chemical vapor deposition (PECVD) on the cathode electrode at high bias (800V) following description of reference [7]. The films were deposited at room temperature using a flow of methane (CH₄) and keeping the pressure constant at 1.0Pa.

The measurement of the stress was performed by the thermally induced bending (TIB) technique, which comprise of the deflection a double beam laser (created with two mirrors) from the surface of the film. This procedure allows the determination of the curvature of the film/substrate sample from which one can obtain the stress of the film. By making the measurement as a function of temperature one also determines the thermal expansion coefficient of the film. Detailed description is found in reference [3]. The hardness of the films deposited on a silicon substrate was measured using a Berkovich diamond tip from a NanoTest-100 system and adopting the Oliver and Pharr method [8].

III. THEORY

The stress (s) of the films were determined using the bending beam technique, which consist in determining the curvature of a film/substrate composite before and after the film deposition using the Stoney equation [9,10]:

$$s = \left(\frac{E_s}{1-\nu_s}\right)t_s^2/6t_f\left(\frac{1}{R} - \frac{1}{R_0}\right) \quad (1)$$

where E_s , ν_s and t_s are the Young's modulus, Poisson's ratio and thickness of the substrate, respectively, R and R_0 are the radius of curvature of the film/substrate composite and the blank substrate, respectively, and t_f is the film thickness.

For the determination of the coefficient of thermal expansion one needs to perform measurements of the stress as a function of temperature, as illustrated in Figure 1a. The slope of the stress with temperature is related to the thermal expansion coefficient of the film (α_f) and substrate (α_s) by the equation [9,10]:

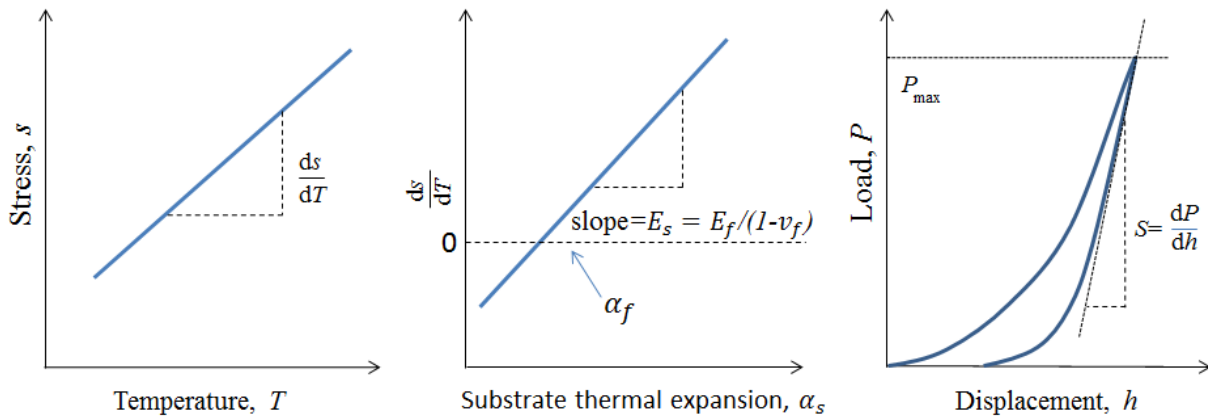


Fig.1: Schematic diagrams of (a) stress (s) of thin film as a function of temperature, (b) variation of stress with temperature (ds/dT) as a function of the coefficient of thermal expansion of the substrate (α_s) and (c) load as a function of tip displacement in typical nanoindentation measurements.

$$\frac{ds}{dT} = \left(\frac{E_f}{1-\nu_f}\right)(\alpha_s - \alpha_f) \quad (2)$$

where E_f and ν_f are respectively the Young's modulus and Poisson's ratio of film. Thus, if one performs measurements of stress as a function of temperature in different substrates, eq. 2 shows that the ds/dT versus α_s is a linear relation, where the intersection with the abscissa axis is the coefficient of thermal expansion (α_f) and the slope (E_s) is the biaxial elastic modulus of the films, i.e.:

$$E_s = \frac{d(\frac{ds}{dT})}{d\alpha_s} = E_f/(1 - \nu_f) \quad (3)$$

The hardness of thin films has been usually determined using nanoindentation techniques with Berkovich diamond tips and adopting the Oliver and Pharr model [8]. Figure 1c displays a sketch showing the main characteristics of the indentation process. A load placed on the indenter is measured as a function of the tip penetration (displacement). The final load is determined by the user and kept constant for a short period and then the indenter is unloaded. The hardness, H , is then determined by:

$$H = \frac{P_{max}}{A} \quad (4)$$

where P_{max} is the maximum load applied to the film and A is the indented area of the diamond tip. The measurement also allows the determination of the elastic constant by using the slope (stiffness, S) extracted from the upper part of the unloading curve, which is given by [8]:

$$S = \frac{dP}{dh} = \frac{2}{\sqrt{\pi}} E_r \sqrt{A} \quad (5)$$

where E_r is the reduced modulus given by:

$$\frac{1}{E_r} = \frac{(1-\nu_f^2)}{E_f} + \frac{(1-\nu_i^2)}{E_i} \quad (6)$$

Where E_i and ν_i are the Young's modulus and Poisson's ratio of the indenter (diamond tip).

Since the properties of the diamond tip are known and the area can be determined by the tip geometry and indentation depth, one can obtain the reduced modulus determined by the nanoindentation process, E_H , as [6]:

$$\frac{1}{E_H} = \frac{(1-\nu_f^2)}{E_f} = \frac{1}{E_r} - \frac{(1-\nu_i^2)}{E_i} \quad (7)$$

or

$$E_H = \frac{E_f}{(1-\nu_f^2)} \quad (8)$$

In summary, from stress as a function of temperature and nanohardness measurements one obtains 5 thermal and elastic properties of thin films: stress (s), hardness (H), coefficient of thermal expansion (α_f), and two reduced elastic modulus, one obtained from stress vs. temperature measurements ($E_s = E_f/(1 - \nu_f)$, eq. (3) and the second from hardness measurement ($E_H = E_f/(1 - \nu_f^2)$, eq. (8). Note that both reduced elastic modulus are different in the exponent of the Poisson's ratio, which is 1 for the reduced modulus obtained from stress and 2 for hardness measurement. That is an interesting result since the two equations are independents, so they allow us to extract separately both the Young's modulus (E_f) and the Poisson's ratio (ν_f) of the film by solving the system of equations (3) and (8). A similar procedure for determining separately the Young's modulus and Poisson's ratio have also been proposed by Ferrari *et al.* [11], but using Brillouin scattering in place of the bending beam technique used here.

IV. RESULTS AND DISCUSION

The above procedure was adopted to determine the thermal and elastic properties of diamond-like (DLC) film. Figure 2 displays the stress (s) as a function of temperature of a DLC film deposited in 2 different substrates: Precision glass 211 and Corning glass 7059. The negative value of the stress indicates the stress is compressive, which has usually been observed in amorphous carbon film [5,6]. The value is relatively low since the film was prepared at high bias producing a material with high concentration of sp^2 bonds. The positive slope (ds/dT) of those curves indicates that the thermal expansion coefficient of the film is smaller than those of the substrates, and the higher the difference the higher the slope.

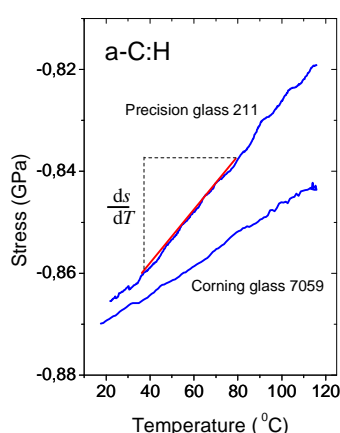


Fig.2: Stress as a function of temperature of a-C:H films deposited in two different substrates (Precision glass 211 and Corning glass 7059).

The slope of the measurements displayed in Fig. 2 can be plotted as a function of the coefficient of thermal expansion of the substrates. In Fig. 3 we show an example of this procedure using a-C:H films deposited on 5 different substrates (silicon and germanium (111), sapphire, Precision glass 211 and Corning glass 7059). One can observe that it follows a linear relation with the coefficient of thermal expansion of the substrate. A linear fitting of the data gives a slope ($E_s = d(ds/dT)/d\alpha_s = E_f/(1 - \nu_f)$) equal to 95 GPa, and the intersection with the abscissa gives the coefficient of thermal expansion of the film (α_f) equal to $2.2 \times 10^{-6} \text{ } ^\circ\text{C}^{-1}$, according to equation (2).

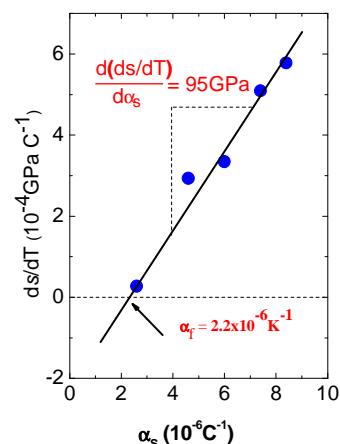


Fig.3: Slope of stress versus temperature as a function of the coefficient of thermal expansion of the substrates

Table 1 presents the results obtained for the stress and hardness measurements of a-CH films deposited on (111) silicon substrates. The reduced biaxial modulus determined by stress was $E_s = 95 \text{ GPa}$ (Fig. 2), whereas the reduced elastic modulus determined by nanoindentation was $E_H = 80 \text{ GPa}$. The difference is due to the fact that these reduced elastic moduli are different. By adopting the procedure described in the theory section, one can use this difference to extract separately the pure Young's modulus ($E_f = 77 \text{ GPa}$) and the Poisson's ratio ($\nu_f = 0.19$) of the a-CH film using equations (3) and (8).

Table.1: Thermal and elastic properties determined through stress as a function of temperature and nanoindentation and using the procedure described in the text.

Stress	0.87 GPa
Hardness	14 GPa
Thermal expansion coefficient	$2.2 \times 10^{-6} \text{ } ^\circ\text{C}^{-1}$
Biaxial modulus, $E/(1-\nu)$	95 GPa
Reduced modulus, $E/(1-\nu^2)$	80 GPa
Young's modulus	77 GPa
Poisson's ratio	0.19

V. CONCLUSION

Measurements of stress as a function of temperature of thin films deposited on substrates with different coefficient of thermal expansion supply an elastic modulus different from the one obtained by nanoindentation measurements. In this work we present a procedure using these two techniques to obtain separately the Young's modulus and the Poisson's ratio of thin films. Thus, by adopting these two techniques one can determine 5 thermal and elastic properties of thin films:

stress, hardness, coefficient of thermal expansion, Young's modulus and Poisson's ratio. The procedure was adopted to obtain those data from hydrogenated diamond-like (DLC) carbon films, a-C:H.

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